

## General comments

In this paper, Li et al. present a new box model that demonstrates high computation efficiency with reasonable accuracy. The model's performance has been thoroughly evaluated through chamber experiments and in-situ observations, showcasing its capability to reproduce pollutant and radical concentrations under varying initial conditions. The results suggest the model could potentially benefit the modeling community. The paper is well-written and adequately referenced. There is only one general comment that I would like to pose. I commend the authors for discussing the implications of their box model in supporting more complex models, as mentioned in the Introduction. I am curious about the potential for easy adoption of the new model in existing CTMs or coupling with climate models. This aspect could significantly enhance the utility of the model if this is the case or it's planned in future developments.

## Response:

Thank you very much for your comments. As of the current version (V1.0), ROMAC operates as a standalone model and does not offer integration capabilities with CTMs. However, in our future development roadmap, we have plans to introduce a modeling framework version of ROMAC known as "ROMAC-plug-in". The ROMAC-plug-in will provide the functionality to be called from Python or Fortran, while preserving its efficient design. This kernel will empower users to seamlessly construct their own models or integrate ROMAC into existing CTMs. We have included this development plan in the future development section in the revised manuscript.

Line412-416: In future development roadmap, we have plans to introduce a modeling framework version of ROMAC known as "ROMAC-plug-in". This ROMAC-plug-in will support calls from Python or Fortran, ensuring compatibility and flexibility for users. Importantly, the efficient design of ROMAC will be maintained, allowing for optimized performance. The kernel of ROMAC-plug-in will be specifically engineered to provide users with flexibility to effortlessly construct their own models or integrate ROMAC with existing frameworks, such as CTMs.

### Specific comments

1. Eq.1: It is essential to address why wet deposition is not included as a default item in the function, especially for hydrophilic components like sulfate. Providing an explanation or a discussion on this matter would add clarity to the model's capabilities.

### Response:

Thank you for your valuable suggestion. The primary focus of our box model simulation centers on chemical processes. In prior studies, the wet deposition process was often overlooked in the photochemical box model. Consequently, we did not develop a specific input interface for it. However, recognizing the significance of wet deposition in certain scenarios, we have included a freely definable rate term denoted as  $(\frac{dc}{dt})_{others}$ . This empowers users to introduce wet deposition into their simulations if deemed necessary. An explanation has been thoughtfully added to Section 2.

Line99-102: Note that the current version of ROMAC does not feature a dedicated input function for wet deposition. Instead, the ROMAC model allows users to set a custom rate term,  $(\frac{dc}{dt})_{others}$ , which can be employed to account for wet deposition. If wet deposition is important for the simulation case, especially concerning the chemical mechanism of hydrophilic components like sulfate, it is suggested that the user incorporates it into  $(\frac{dc}{dt})_{others}$ .

2. Line145: The term "overcome" may not be suitable in describing the model's superiority over existing solvers, as it suggests that the issues present in other solvers have been completely resolved. Instead, consider rephrasing it to highlight that the new model offers an optimized algorithm that strikes a balance between efficiency and accuracy. Maybe also consider to replace the phrase throughout the

paper.

**Response:**

Thank you for pointing out the improper application of words in the manuscript. We have used the term “outperform” to replace “overcome” in the revised manuscript. This sentence has been modified to emphasize the advantages of the VSVOR solver over the EBI solver:

Line171: Hence, this scheme will enhance the applicability and stability of the ROMAC numerical solver compared to the EBI numerical solver.

We modified the sentences that used the word 'overcome' throughout the paper.

Line14-16: ROMAC outperforms the traditional box models in evaluating the impact of physical processes on pollutant concentrations, and its ability to quantify the effects of chemical and physical processes on pollutant concentrations has been confirmed by the chamber and field observation cases.

Line84-85: Therefore, ROMAC will be computationally efficient and outperform the traditional box models in evaluating the impact of physical processes on pollutant concentrations.

Line391-392: Compared with the traditional Observation Based box Model (OBM), ROMAC is superior in evaluating the impact of physical processes on pollutant concentrations.

3. Line184-185: ‘the VSVOR solver has comparable computational efficiency with the EBI solver, and the solution accuracy and stability are better’ – any obvious evidence on this than the equations listed above?

**Response:**

Thanks for the comment. Yes, in addition to the equations, other evidences include a series of numerical experiments conducted to test the accuracy and stability of EBI and VSVOR which are presented in Section 2.4.

In terms of accuracy, in general, the VSVOR solver has second-order accuracy, and theoretically possesses a smaller truncation error compared to that of the EBI solver. In addition, we also evaluated the accuracy of the results by comparing with the results of AtChem (Figure 3). The differences between the VSVOR solver and AtChem results are all within the preset relative tolerance ( $10^{-3}$ ). This is because that the VSVOR solver has a smaller truncation error and also has a strict error control scheme. However, the EBI solution results of some species (*e.g.*, NO, NO<sub>2</sub>, MGLYOX) will exceed the preset relative tolerance.

In terms of stability, the fixed-step EBI solver may not converge due to the preset integration time step size being too large, which can be known from the test results in Table A2. The VSVOR solver with an adaptive variable time step size scheme can find the optimal integration time step and operating in a stable manner.

To reduce confusion, we have moved this conclusive statement to the end of Section 2.4 so that the readers can combine theoretical and numerical experimental results for a better understanding of the advances of the VSVOR solver.

Line 310-312: However, reducing the integration time step too much diminishes the efficiency of the EBI solver when handling the MCM mechanism in comparison to the VSVOR solver. Hence, the VSVOR solver exhibits comparable computational efficiency to the EBI solver, while maintaining superior solution accuracy and stability.

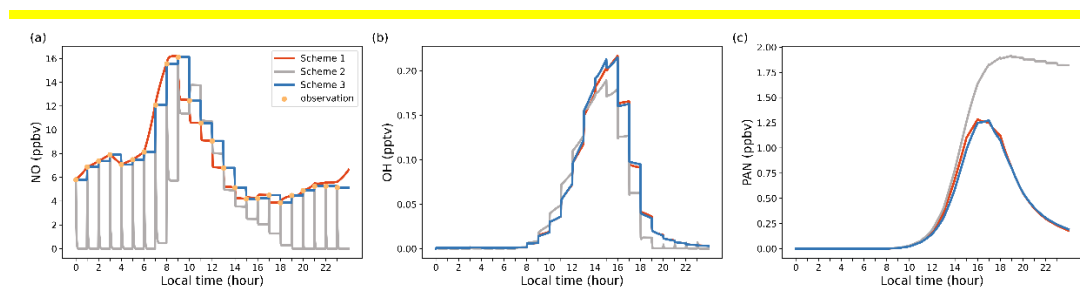
4. L232-233: It would be beneficial to offer a general recommendation on the choice of scheme for commonly studied species (*e.g.*, O<sub>3</sub>, PAN, SO<sub>2</sub>) when utilizing the model. Users might find such a guide helpful when first implementing the model.

**Response:**

Thanks for your suggestion. We have incorporated the simulation results for PAN into Figure 2c. This inclusion provides reader with insight into how varying precursor constraint schemes can alter the simulation results for secondary pollutants. Users are

encouraged to select the most suitable scheme in accordance with their requirements. The scenarios in which each scheme can be applied have been described in Section 2.2. Additionally, we have rectified an error in the OH radical results that stemmed from a previous data processing mistake.

Line257-260: It is worth noting that due to variations in constraint schemes, simulated concentrations of other species, such as OH and PAN, can also diverge (Figure 2b and 2c). This case study was primarily designed to elucidate the unique features of different constraint schemes, with no intent to definitively validate or invalidate any particular scheme. Users are encouraged to make their scheme selections judiciously, aligning them with their specific research needs and observational findings.



**Figure 2. Model output results illustrating diurnal variations for selected species, highlighting the impact of different concentration constraint schemes. (a) NO concentrations; (b) OH concentrations; (c) PAN concentrations.**

5. The model description section employs numerous abbreviations, which may hinder readability. I recommend creating a table containing all abbreviations to enhance the section's clarity and ease of understanding.

**Response:**

Thank you for your valuable suggestion. To enhance the manuscript's readability, we have compiled a comprehensive list of abbreviations along with their corresponding descriptions in Table B1, which is included as an appendix.

Line107-108: The subsequent section offers a comprehensive overview of ROMAC's

features. Furthermore, to facilitate reference, all parameters employed in this paper are cataloged in Table B1.

**Table B1** Nomenclature

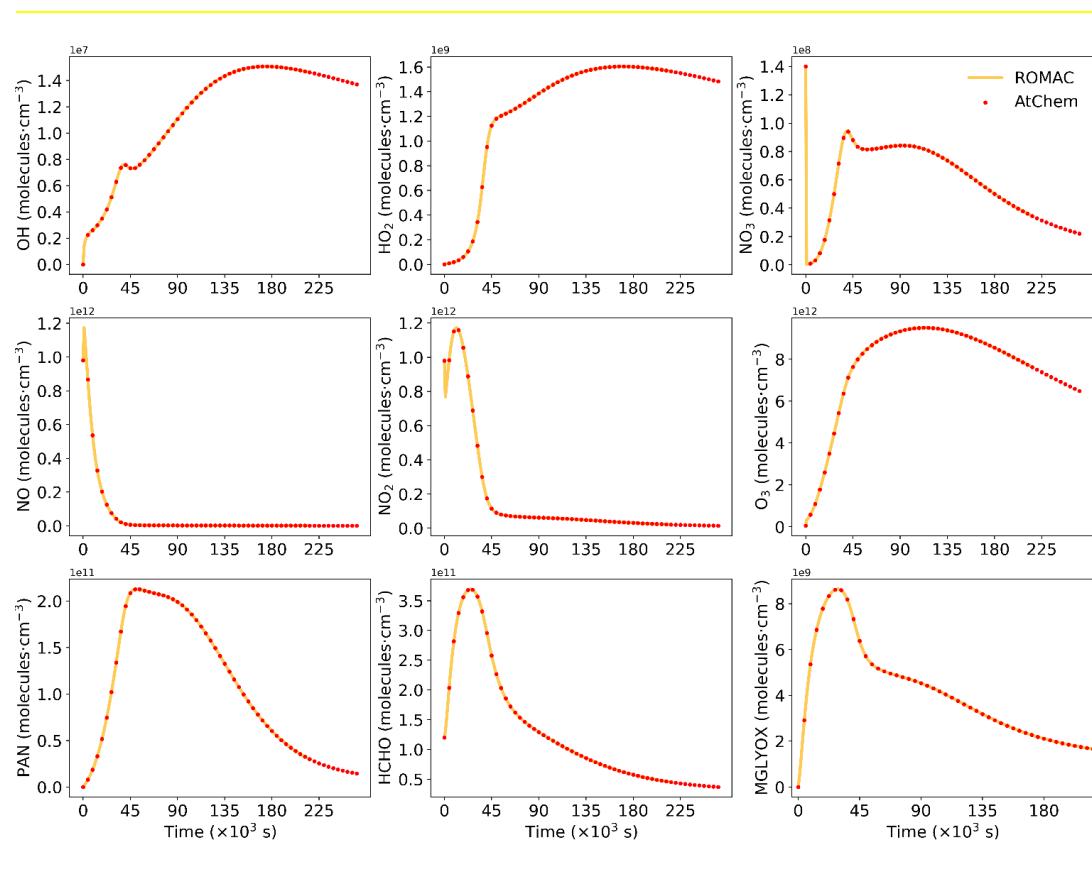
Abbreviations	Explanation
ODEs	Ordinary Differential Equations
VSVOR	The variable-step and variable-order solver
$atol$	absolute tolerance
$rtol$	relative tolerance
$r$	The reactant in a chemical reaction
$p$	The product in a chemical reaction
$\alpha, \beta$	Stoichiometric number
$C_t$	Concentration of species at time $t$
$f_i(C_{i,t}, t)$	Rate of change of species $i$ at time $t$
$P_{i,t}$	Product rate of species $i$ at time $t$
$L_{i,t}$	Loss rate of species $i$ at time $t$
$l_{i,t,R}$	The part of the chemical reaction rate that is not directly related to the concentration of species $i$ in reaction $R$ at time $t$
$\Delta t$	Integration time step size
$g_1(C_{t+1})$	The objective function when Newton's method solves the implicit Euler method
$g_2(C_{t+1})$	The objective function when Newton's method solves the implicit trapezoidal method
$C_{t+1}^k$	Species concentration at iteration $k$ of Newton's method
$\nabla g_1(C_{t+1})$	The Jacobian matrix of $g_1(C_{t+1})$
$\nabla g_2(C_{t+1})$	The Jacobian matrix of $g_2(C_{t+1})$
$\nabla g^{-1}(C_{t+1})$	The inverse of the Jacobian matrix
$\Delta t_0$	Integration time step size equal to $2.22 \times 10^{-16}$ s
$\Delta t_1$	Minimum specie atmospheric lifetime in chemical mechanisms
$\Delta t_2$	The maximum time step size necessary to achieve diagonal dominance of the Jacobian matrix.
$\Delta t_{init}$	Initial integration time step size
$\Delta t_{max}$	The maximum integration time step to ensure the result does not exceed the preset tolerance
$\Delta t_{opt}$	Optimal integration step size
RERR	Relative error calculated by the doubled-step method
LTE	Local truncation error
$atol$	Absolute tolerance
$rtol$	Relative tolerance
$Rn$	Lagrangian remainder in the Taylor expansion
$\xi$	Real number in the Lagrangian remainder in the Taylor expansion
RMSE	Root Mean Square Error

6. Figure A1: To improve clarity, consider using more distinct colors for the two models or converting one model to a scatter plot. Which solver is used to obtain the ROMAC results?

**Response:**

Thanks for your suggestion. We have now altered the representation of AtChem results to a dot shape. Also, the solver used by ROMAC in this test has been given the revised manuscript and also in the figure's caption.

Line298-299: The simulation results for ROMAC in Figure A1 are processed by the VSVOR solver.



**Figure A1. Comparison of the simulation results between ROMAC and AtChem for nine substances. ROMAC used the VSVOR solver in this test.**

7. Line 285: The subtitle may not be suitable: it includes both model evaluation (esp. the chamber study section) than application.

**Response:**

We appreciate your comment, and we have made the following adjustments accordingly:

- The subtitle of Section 3 has been modified to "Model Validation and Application."
- The subtitle of Section 3.1 has been updated to "Chamber Simulation Case."
- The subtitle of Section 3.2 has been revised to "Field Observation Case."

8. Line 318-319: I'm not sure if such a conclusion can be drawn from Fig. 4c, as significant uncertainty exists in  $k_{other}$ .

**Response:**

Thank you very much for pointing out this issue. The misleading statement, "Based on dynamic optimization, ROMAC can overcome the shortcomings of the over-simplified physical process in the traditional box model.", has been removed in the revised manuscript.

In this simulation case, the main reason that the observation cannot be reproduced in scenario 1 is that the physical process is missing in the model. This view can be proved by the fact that the simulation results better match the observation results after adding the theoretical calculation to the physical process in scenario 2. However, it is worth noting that there are still gaps between scenario 2 and the observations. Therefore, there should be uncertainty in the estimation of this physical process. But the expected value should be consistent with the theoretical calculated value. Dynamic algorithms incorporate fluctuations that cannot be captured by theoretical calculations into the results when calculating the effects of physical processes. Hence, the  $k_{other}$  in scenario



3 have a range of fluctuations, however the average value is close to the theoretical calculation can prove that this scheme is feasible. A note on uncertainty has been added to the manuscript:

Line350-354: The rate of the physical process is subject to uncertainty in practical applications, but its average value is expected to closely approximate the theoretical value. The optimized value of  $k_{others}$  in scenario 3, as shown in Figure 4c, exhibits a certain range of fluctuations rather than a fixed value. However, its average values ( $1.430 \times 10^{-5}$ ) are comparable to  $k_{dilu}$  in scenario 2 (Figure 4c), which indicates that the dynamically optimized algorithm is reliable.